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Historical Releases of Radioactivity to the Environment from ORNL

W. F. Ohnesorge

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prepared by

W. F. Ohnesorge

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HISTORICAL RELEASES OF RADIOACTIVITY TO THE ENVIRONMENT FROM ORNL

1. INTRODUCTION

The purpose of this report is to give a brief history and assessment of ORNL radionuclide releases to the environment. A short history and an inventory of radioactivity disposed of by shallow land burial and hydrofracture techniques are given along with a brief discussion of the potential environmental impact of these disposed materials. The data available for this report varied greatly in quality. Data on contaminated liquid waste and liquids discharged to the environment are much more reliable than early data on contaminated air discharges and contaminated solid waste. Data for more recent years are more complete and reliable than data obtained from records dating back to the early history of the Laboratory. The data presented here do not include materials retrievably stored (with the exception of uranium solid waste data) or inventories of materials stored in operating or surplus facilities since these materials presently have no contact with or loss to the environment.

2. RADIONUCLIDE RELEASES VIA THE WATER PATHWAY

2.1 Introduction

Currently, the most significant radionuclide released by the water pathway is strontium-90 (Sr-90) because of its radiotoxicity, environmental availability, and the quantities released. Other significant radionuclides are tritium, cesium-137 and transuranics (long-lived alpha emitters with atomic number greater than 92).

Presently there are three major source categories of radionuclide-contaminated releases from ORNL through the liquid pathway to the environment. Prior to 1985, the primary source (on the order of 70 to 80 percent) of Sr-90 releases was the leaching of radionuclides from the waste disposal areas to White Oak Creek or Melton Branch. White Oak Creek and Melton Branch converge and flow into White Oak Lake which empties into the Clinch River at Clinch River Mile 20.8. Because of problems with liquid waste systems in the main ORNL complex, a large portion of the three curies of Sr-90 over White Oak Dam in 1985 came from sources other than waste disposal areas.

Another source (normally, on the order of 10 percent of all Sr-90) is waste water from operating facilities such as research reactors, chemical processing plants and research laboratories. These process waters either go directly into White Oak Creek or to a temporary hold-up basin for testing and treatment (if needed) before release to the creek. Due to improvements in treatment of process water, only a small fraction of present radionuclide discharges over White Oak Dam are due to this source.

In addition to the above sources of radionuclides in water, there are discharges (on the order of 10 percent of all Sr-90, with the exception of 1985) due to leaching of materials from contaminated surfaces and soils in the area of the operating facilities. The origins of this contamination are from previous spills, and leaking underground pipes and tanks. The pathway to White Oak Creek is either by way of storm water runoff or cross contamination between liquid waste and drain system pipes. A determined effort is being made to reduce this source of discharge.

2.2 History of Radionuclide Releases to the Water Pathway

During the years from 1943 to 1949, liquid waste treatment consisted of holding the liquid in tanks for radioactive decay, then decanting

to settling basins for further radioactive decay and separation of particulate matter before discharge to White Oak Lake. Addition of sodium hydroxide prior to discharge caused most of the Sr-90 to be precipitated. White Oak Lake provided additional settling and decay time prior to discharge to the Clinch River (Browder 1959).

From 1949 until 1954, the more highly radioactive liquid waste was concentrated in an evaporator and stored in concrete tanks. Starting in 1951, much of the liquid waste was placed in pits and trenches (see Tables 1 and 2). The evaporator was taken out of service in 1954 and, subsequently, all of the liquid radioactive waste went to pits and trenches until 1963.

In 1950 the Laboratory was greatly expanded in size and its scope of operations. The liquid waste systems were expanded and improved by the addition of tanks, monitoring systems and segregation of liquid wastes. In 1954, a pump and a 1 1/2 mile pipeline from the collection tank area to the disposal pit area were installed to replace a tank truck that had previously been used to transport liquid waste. In 1957 a waste water treatment plant (a lime-soda process) was put into operation. This was to reduce the level of radioactivity in the low activity process waste water discharged to White Oak Creek. Also in 1957, a fission product pilot plant was constructed to recover Sr-90, Cs-137, and other valuable radioisotopes. The isotopes thus recovered were prevented from entering the environment by being solidified and securely contained.

During 1957 when large amounts of Ru-106 (ruthenium-106) were discharged to the waste pits, noticeable seepage was found coming from one pit. Much of this seepage made its way into White Oak Lake as may be seen in column 4 of Table 3. Even though discharge of Ru-106 to the pits was greatly reduced after 1959, seepage of that radionuclide was evident for a number of years afterward. All liquid discharges to the pits and trenches were discontinued in

1966, although small amounts of sludges continued to be placed in the pits until 1976 (See Table 1).

In 1964 a hydrofracture facility was put into service. A new waste evaporator was built to concentrate the low level liquid waste. Liquid radioactive waste concentrate was mixed with a cement grout mixture and injected deep into the earth to minimize its release to the environment. As can be seen from Table 4, material was processed in this facility until 1979. A second hydrofracture facility (later called the Shale Fracture Facility) was placed into service during 1982 and most of the remaining liquid radioactive waste concentrate and sludge waste that had been stored in gunite tanks was disposed during the period 1982 to 1984 (Table 5). The Shale Fracture Facility has not operated since January 1984. Recently a question of possible leaching to deep groundwater has been raised and no more disposal by this method will occur until further studies have been completed.

During 1976 an improved process waste treatment plant was put into service. This plant greatly reduced the amount of radioactivity being discharged from process water to White Oak Creek. From 1976 to the present there has been a continuing series of projects to improve the liquid and solid waste systems so that radioactive discharges from White Oak Lake could be reduced. Some of these include rerouting of water around waste burial areas and putting caps over some of the trenches. Installation of linings in the sanitary sewer system was recently completed which should reduce the amount of radioactivity getting into White Oak Creek through the Sewage Treatment plant.

2.3 Liquid Waste Data

The liquid waste data are based upon the radiochemical analyses of samples taken from each batch of liquid disposed and should be a reasonable estimate of the quantities of the various radionuclides in the waste. Tables 1 and 2 show radioactive liquid and sludge disposed of in pits and trenches. Tables 4 and 5 show liquid disposal to the shale fracture facility. While disposal records show most of the radioactivity to the pits as "unidentified beta-gamma" for the years 1951 through 1960, one reference (Lomenick et al. 1967) gives the following breakdown: 42,000 Ci Sr-90, 182,000 Ci Cs-137 (cesium-137), 230,000 Ci Ru-106 and 70,000 Ci of trivalent rare earths (exclusive of yttrium-90). For the years 1963 through 1976, the data represent an estimate of radionuclides in process waste treatment plant sludge which was disposed of in Pit 4. In Tables 1 and 2, the heading "TRE" represents total rare earths. These include radioactive isotopes of the elements lanthanum, cerium, praseodymium, neodymium and promethium. The longest lived of the more significant radionuclides in this group is promethium-147 which has a half-life of 2.6 years.

In summary, an estimated total of 600,000 curies of Cs-137 and 200,000 curies of Sr-90 were disposed of in the pits and trenches. Based on the half-lives of these nuclides which are on the order of 30 years, there are an estimated 400,000 curies of Cs-137 and 120,000 curies of Sr-90 still present in the pits and trenches area. Due to their short half-lives, most of the Co-60 (cobalt-60), Ru-106 and TRE have decayed. The nuclides that remain represent a potential for environmental releases. Studies are being performed to determine what, if any, action might be required to prevent significant future releases.

On the order of 700,000 curies each of Sr-90 and Cs-137 were disposed of in the shale fracture facilities. There still remains

on the order of 600,000 curies of Sr-90 and 500,000 curies of Cs-137 after radioactive decay. For many years it was thought that the shale fracture method of disposal was the ultimate method for ORNL's liquid radioactive waste and that materials disposed in this way would not likely pose any environmental concern. As previously mentioned, a question of possible leaching to deep groundwater has been raised. No more injections will occur unless all legal and technical issues are resolved.

2.4 Solid Waste Disposal Data

The solid radioactively contaminated waste (disposed by shallow land burial) disposal data are given in Tables 6 and 7. Many of these data should be considered to be only crude estimates of the radionuclides buried as solid waste. A portion of the uranium shown in Table 7 is in retrievable storage. Although records of waste volume were maintained, no detailed records of radioactivity in solid waste disposal were kept prior to 1977. Some of the burial ground records were inadvertently destroyed by fire. Estimated values of radionuclides disposed before 1977 are based on volume records over various integrated time periods, best estimates from individuals who participated in the programs, and accountability records. Due to the uncertainty of these early data, it is not presented by year (Table 6). Since uranium data are based on accountability records, it is thought to be more reliable and is therefore presented by year in Table 7. The accountability records did not distinguish between buried and retreivable stored uranium, therefore, both are included in Table 7.

For 1977 and later, data are thought to be much improved over previous years, but there are still uncertainties due to difficulty in quantifying the amount of contamination on solid waste

materials. Many of the values furnished on waste disposal forms are estimates given as conservative (estimate larger than actual) less-than values which are entered into the computer data base as "actual" values.

2.5 Trends of Discharges Over White Oak Dam (WOD)

Table 3 shows the quantities of radionuclides discharged to the Clinch River for the period 1944 through 1985. No data for individual nuclides are available for the earlier years, 1944-1948. These data were reported as gross-beta and were considered to be crude estimates. Gross beta was measured daily using direct radiation readings. After 1948, radiochemical analyses were made to determine the species and quantities of radionuclide releases (Cowser and Snyder 1966). One of the greatest uncertainties in data was the uncertainty in flow data at high flow rates. In 1984 weirs were completed at White Oak Creek, Melton Branch and White Oak Lake to provide more accurate flow measurements over a wide range of flows. As previously mentioned, the large amounts of Ru-106 going over White Oak Dam starting in 1959 were due to leakage from a liquid waste disposal pit. The decrease of ruthenium over WOD after 1961 was roughly proportional to the radioactive half-life of the isotope (368 days). The decreases in Cs-137, Sr-90 and other radionuclides over the dam after 1958 were at least partly due to increased efficiency of waste-water treatment. Most of the radioactive discharges (with the exception of 1985 discharges) have been due to leaching from the waste disposal areas.

2.6 Water Pathway Dose Calculations

2.6.1 Drinking Water

Population dose commitments were not calculated for the period 1944 to 1948 since these calculations are nuclide specific, and only data

based on crude estimates of gross beta are available. For the period 1949 through 1964, Sr-90 and Ru-106 were the major contributors to dose commitment. From 1965 to 1985 Sr-90, tritium (H-3), and alpha emitters were the major contributors.

The population dose estimates are based upon individuals obtaining all of their required water intake from the Tennessee and Clinch Rivers. No credit was taken for reduction of concentration due to water treatment or natural means such as sedimentation. The population included all people who obtain water from public water supplies for approximately 100 miles downstream from ORNL. Also, the K-25 worker population was included. For the total reporting period (1949-1984), the total population dose commitment was on the order of 3000 person-rem. This should be compared with natural background radiation which contributed on the order of 9,000,000 person-rem to the same population over the same period of time.

The estimated maximum dose to any organ of any individual due to drinking water during any year was assumed to be to an individual worker at K-25 who drank one liter of water each working day for 250 days in a year. The calculated maximum annual dose commitment to any organ was 30 mrem to the lower large intestine. This result was obtained for each of the years 1960 and 1961, and was primarily due to Ru-106. For the past 11 years the estimated maximum dose to any organ of a K-25 worker by the same pathway has been less than 1 mrem per year.

2.6.2 Fish Consumption

During the period 1960 to 1962, a special study was performed to determine the dose commitments due to eating fish taken from the Clinch and Tennessee Rivers (Cowser and Snyder 1966). This was at a time when discharges of Ru-106 from ORNL were highest but the discharges of Sr-90 were averaging on the order of 14% of the

highest annual discharges which had previously occurred. The results indicated that Sr-90 was the nuclide which contributed most of the dose commitment to any organ. The average dose commitment due to eating 37 lbs (assumed annual consumption) of fish from the Tennessee River was approximately 3 mrem to the bone (endosteal cells) and 0.3 mrem effective total body dose equivalent. This estimate assumes that no fish bones were consumed. A population dose due to eating all of the fish harvested from Watts Bar during one year (approximately 610,000 pounds) was estimated to be on the order of 5 person-rem effective total body dose equivalent.

Data for fish taken from the Clinch River during the same 1960 to 1962 period indicated that the maximum dose to any organ was 7 mrem to the bone (endosteal cells) if no fish bones were consumed. Data indicated that if bones were included in the consumed fish, the maximum dose to the individuals bone would have been on the order of 30 mrem. During the time of the referenced study, DOE (AEC) Manual Chapter 0524 was the applicable regulatory guide and it specified that a member of the public should not receive more than 500 mrem to the whole body, gonads or bone marrow; or 1500 mrem to any other organ in any year as a result of AEC sponsored operations.

The referenced study gave no indication of exactly where on the Tennessee and Clinch Rivers the fish were caught. Other data indicate that the location of the catch on the Clinch River can influence the radionuclide concentrations. For example, in 1984 Sr-90 in carp caught at Clinch River Mile 20.8 (mouth of White Oak Creek) was at least 5 times higher than in those caught elsewhere on the Clinch. The dose due to eating 37 lbs of these carp would have been on the order of one mrem to the bone (endosteal cells) of the individual if only carp flesh were eaten (fine bones included but no large bones such as rib or back bones). If the large bones were ground in with the flesh to make fish patties, the dose commitment due to eating each pound of fish patty would be approximately 7 mrem to the individual's bone (endosteal cells).

3. ENVIRONMENTAL RELEASES OF RADIONUCLIDES TO AIR

3.1 Routine Unmonitored Releases

Prior to 1950, discharges to air were from stacks serving individual facilities. The two most significant discharges to the air were from the stacks serving the RaLa (radioactive lanthanum) facility and the Graphite Reactor. The RaLa facility was a major processor of short-cooled reactor fuel slugs for the production of radioactive lanthanum. This facility operated from 1945 to 1956, and until early 1949 there was no gas discharge treatment. Particulates, noble gases and radio-iodines were released in unknown amounts. I-131 (iodine-131) was probably the most significant radionuclide released because of the short decay of the reactor slugs prior to processing, and because of the volatility and radiotoxicity of I-131. There was no routine sampling and reporting of I-131 discharges, but the results of a special study indicated the discharge of curie quantities during one RaLa run (Winters 1948). One report (Davis 1949) indicated that 1200 Ci of I-131 was included in the radioactive inventory of each run, but that filters and scrubbers of unknown efficiency were used to treat off-gases before discharge to the stack.

From 1944 until 1948, the graphite reactor was operated without a filter system. No significant particle discharge was noted from this facility until 1947 when a series of slugs in one reactor channel ruptured and an attempt was made to clear the channel of these slugs. After this time and until the filter system was installed in 1948, activity could be detected in the streets of the Laboratory on a regular basis (McLain, 1948).

The graphite reactor also discharged argon-41 while operating. This was due to neutron activation of natural argon in the cooling air. An estimate of 400 curies discharged per day was given in one report (Browder 1959).

The central off-gas and ventilation system with a 250 foot stack (Stack 3039) was built about 1950. Particulate filters and an electrostatic precipitator were included at the time of installation, but no scrubber or other system was included to trap the radioiodines. Scrubbers were first installed in the central system in early 1961.

The earliest routine discharge data available are for I-131 discharged in 1961. Data on particulates were also obtained by this time, but the quantities discharged have been insignificant compared with the gaseous discharges. A much improved sampling system was installed the following year (1962). In 1970 the stack sampling system was further updated and noble gas discharges were reported for the first time. Also during 1970, charcoal filtering was added to the off-gas line of a liquid waste storage tank used to store waste which came from an I-131 production process.

Tritium discharges and unidentified alpha particulates were first reported in 1972. An idea of the possible range of discharges of tritium to the atmosphere prior to discharge records may be obtained by examining the isotope sales records for sales of tritium. During the first reporting year for tritium discharges out the central stack system, the discharges were reported at 1800 curies. During that year sales records indicated sales of 161,000 curies. During the years 1965 through 1971, annual sales varied from 121,000 curies to 334,000 curies. Total sales prior to 1965 were 363,000 curies. Noble gas releases were not monitored until 1970. The noble gases are reported as krypton-85 and xenon-133 in Table 8. The major sources of noble gas releases are the Oak Ridge Research Reactor

(ORR), the High Flux Isotope Reactor (HFIR), and processing of fissionable materials. The ORR has been in operation since 1958 and the HFIR since 1965. Since reporting began in 1970, noble gas discharges have been relatively constant. Another reactor facility which was operational prior to noble gas discharge reporting was the Low Intensity Test Reactor (LITR) which operated from 1951 to 1968.

In addition to the more or less routine unmonitored releases described above, there were several incidents which caused additional unmonitored discharges. The two most significant of these events were a ruthenium release incident and an incident at Building 3019.

3.2 Ruthenium Fall-Out Incident

In November 1959, there were two related discharges of large particles contaminated with ruthenium-106 (Borkowski et al. 1960). Health physics survey and stack sample results indicated that the particles were very large and that the contamination was confined to the ORNL area. It was determined that the contamination originated from the main stack after repairs had been made to the blower system. A rough, upper limit estimate of 16 curies was given for the activity on the ground in the vicinity of the stack as a result of the ruthenium-106 releases.

3.3 Building 3019 Incident

A chemical explosion, during 1959, in a hot cell of Building 3019 caused the building containment to be breached and approximately 600 mg of plutonium-239 was spread over a limited area south and east of Building 3019 (King and McCarley 1959). No individual received greater that 2% of the lifetime body burden of plutonium. The estimate of plutonium released was believed to be within an order of magnitude of the actual value at best. The graphite reactor which is located adjacent to Building 3019 was out of service for approximately one month as a result of the incident.

3.4 Reported Releases to the Atmosphere and Their Significance.

Table 8 shows the curies released to the atmosphere during the reporting period. From 1979 to the present, tritium (H-3) has been the primary contributor to the population dose commitment. The increasing amounts of tritium discharged is a reflection of the greatly increased processing associated with sales of this isotope. In years 1972 to 1979, tritium and iodine were roughly equal in significance. Prior to 1972, I-131 was probably the most significant nuclide released to the atmosphere. The decrease of I-131 release with time has been due to decreased production and better off-gas treatment.

The 50 mile population dose due to the reported discharges to air for the period 1961 through 1984 was on the order of 900 person-rem. This can be compared with a population dose of 3.5 million person-rem due to natural background radiation to the same population over the same period of time. The 30 mile population dose was calculated to be on the order of 800 person-rem due to ORNL operations. This can be compared with 2.6 million person-rem due to natural background radiation to the population within a 30 mile radius of ORNL.

When iodine was the most significant discharge, the thyroid was the organ receiving the largest amount of radiation. For 1962, the year when the largest iodine discharge was reported, calculations indicate the maximum dose to any adult individual's thyroid was on the order of 30 mrem. During the last 5 years, estimated maximum organ and total body doses to any individual member of the public have been less than 1 mrem which may be compared with current EPA guidelines of 25 mrem per year to the total body and 75 mrem per year to the critical organ.

4. SUMMARY

A brief history of radioactive waste disposal and treatment at ORNL and its impact on the environmental pathway has been given. Current practices for handling and treating radioactive waste have improved greatly over those used in the early years of the Laboratory's operation. At the present time, a determined effort is being made to operate with reduced waste volumes while developing future disposal methods.

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Table 1. Curies in Liquid to Pits 1, 2, 3, and 4

Year	SR-90	UID Beta[1]	PU-239	CS-137	CO-60	TRE[2]	RU-106
1951[3]	(4)	390					
1952		953	0.0				
1953		77,165	0.2				
1954		7,224	1.0				
1955		21,390	1.6				
1956		34,990	2.6				
1957		41,920	2.9				
1958		52,790	3.1				
1959		280,000	3.5				
1960		21,490	2.3				
1961	1,671		3.0	12,890		837	757
1962	1,539		1.9	17,560	111	1,141	741
1963[5]	20						
1964	12	37					
1965	6	20					
1966	14	61					
1967	16	42					
1968	9	32					
1969	6	19					
1970	6	17					
1971	6	12					
1972	15	20					
1973	11	13					
1974	4	8					
1975	3	3					
1976	2	2					
TOTAL	3,339	469,150	22.3	30,450	111	1,978	1,498

[1]Unidentified gross beta and gamma emitters.
[2]Total rare earths.
[3]1951 data are for Pit 1. Data for all following years are for Pits 2, 3 and 4.
[4]Blanks indicated that no activity was reported.
[5]Data for 1963 through 1976 are estimated values disposed in sludge. Data for previous dates are for liquid discharges.

Table 2.

Curies in Liquid to Trench 5

Year	SR-90	UID Beta[1]	CS-137	RU-106	CO-60	TRE[2]	PU-239
1960	(3)	3,536					0.8
1961	1,116		13,120	830		41	0.3
1962	1,354		14,750	1,274	153	608	1.7
1963	4,851		48,110	1,096	837		1.7
1964	5,303		58,690	118	110		1.7
1965	80,240		57,435	388	1,827		1.7
1966	2,719		7,471	24	81		0.3
TOTAL	95,583	3,536	199,576	3,730	3,008	649	8.1

Curies in Liquid to Trenches 6 and 7A

Year	SR-90	CS-137	RU-103	RU-106	CO-60	TRE	PU-239
1961[4]	126	665	13	51	24	146	0.1
1962	38	1,588		358	11	6	0.2
1963	5,250	26,300		1,130	365		1.1
1964	10,580	53,400		189	131		1.7
1965	5,534	28,450		53	125		1.3
1966	3,010	4,576		15	136		0.3
TOTAL	24,538	114,979	13	1,796	792	152	4.1

Curies in Liquid to Trench 7B

Year	SR-90	CS-137	RU-106	CO-60	TRE	PU-239
1962	32	1,688	307	9	5	0.2
1963	6,628	25,950	981	385		1.1
1964	6,867	35,880	126	88		1.1
1965	7,327	34,090	54	158		1.1
1966	2,602	4,339	12	12		0.2
TOTAL	23,456	101,927	1,480	652	5	3.8

[1]Unidentified beta gamma emitters.
[2]Total rare earths.
[3]Blanks indicate that no data was reported.
[4]The data for 1961 are for Trench 6. The remaining data in this subtable are for Trench 7A.

Table 3. Annual Discharges of Radionuclides from White Oak Creek to the Clinch River, 1944 to 1985
(Curies)

Year	Gross Beta	^{137}Cs	^{106}Ru	^{89}Sr	^{90}Sr	TRE(-Ce)[a]	^{144}Ce	^{95}Zr	^{95}Nb	^{131}I	^{60}Co	^{3}H	TRU
1944[b]	600												
1945[b]	500												
1946[b]	900												
1947[b]	200												
1948[b]	494												
1949		77	110		150	77	18	180	22	77		NA[c]	0.04[d]
1950		19	23		38	30	NA	15	42	19			0.04
1951		20	18		29	11	NA	5	2	18			0.08
1952		10	15		72	26	23	19	18	20			0.03
1953		6	26		130	110	7	8	4	2			0.08
1954		22	11		140	160	24	14	9	4	NA		0.07
1955		63	31		93	150	85	5	6	7	7		0.25
1956		170	29		100	140	59	12	15	4	46		0.28
1957		89	60		83	110	13	23	7	1	5		0.15
1958		55	42	NA	150	240	30	6	6	8	9		0.08
1959		76	520	0.3	60	94	48	27	30	1	77		0.68
1960		31	1,900	1.9	28	48	27	38	45	5	72		0.19
1961		15	2,000	2.0	22	24	4	20	70	4	31		0.07
1962		6	1,400	1.7	9	11	1	2	8	0.4	14		0.06
1963		4	430	1.0	8	9	2	0.3	0.7	0.4	14		0.17
1964		6	190	0.8	7	13	0.3	0.2	0.1	0.3	15	1,900	0.08
1965		2	69	0.6	3	6	0.1	0.3	0.3	0.2	12	1,200	0.50
1966		2	29	0.9	3	5	0.1	0.7	0.7	0.2	7	3,100	0.16
1967		3	17	0.7	5	9	0.2	0.5	0.5	0.9	3	13,300	1.03
1968		1	5	0.6	3	4	0.03	0.3	0.3	0.3	1	9,700	0.04
1969		1	2	0.3	3	5	0.02	0.2	0.2	0.5	1	12,200	0.20
1970		2	1	0.3	4	5	0.06	0.02	0.02	0.3	1	9,500	0.40
1971		1	0.5	0.2	3	3	0.05	0.01	0.01	0.2	1	8,900	0.05
1972		2	0.5	NA	6	5	0.03	0.01	0.01	0.3	1	10,600	0.07
1973		2	0.7		7	NA	0.02	0.05	0.05	0.5	1	15,000	0.08
1974		1	0.2		6		0.02	0.02	0.02	0.2	0.6	8,600	0.02
1975		0.6	0.3		7		NA	NA	NA	0.3	0.5	11,000	0.02

Table 3. Annual Discharges of Radionuclides from White Oak Creek to the Clinch River, 1944 to 1985
(Continued)

Year	Gross Beta	^{137}Cs	^{106}Ru	^{89}Sr	^{90}Sr	TRE(-Ce)[a]	^{144}Ce	^{95}Zr	^{95}Nb	^{131}I	^{60}Co	^{3}H	TRU
1976		0.2	0.2		5					0.03	0.9	7,400	0.01
1977		0.2	0.2		3					0.03	0.4	6,200	0.03
1978		0.3	0.2		2					0.04	0.4	6,300	0.03
1979		0.2	0.1		2.4					0.04	0.4	7,700	0.03
1980		0.6	0		1.5					0.04	0.4	4,600	0.04
1981		0.2	0.1		1.5					0.04	0.7	2,900	0.04
1982		1.5	0.2		2.7					0.06	1.0	5,400	0.03
1983		1.2	0.2		2.1					0.004	0.3	5,600	0.05
1984		0.6	0.2		2.6					0.05	0.2	6,400	0.03
1985		0.4	0.007		3.0						0.6	3,700	0.008

[a]Total rare earths minus cerium.

[b]Individual radionuclide data not available.

[c]No analysis performed.

[d]Estimated from measurements made during last quarter of 1949.

Table 4. Curies in Liquid to First Shale Fracture Facility

Year	SR-90	CS-134	CS-137	RU-106	CO-60	PU-238	PU-239	CM[1]	UN-ID Alpha[2]
1964	610	(3)	317	36	4				
1965	822		4,920	4	15				
1966	3		19,950	21	8				
1967	10,050		75,500	594	642				
1968	4,800		121,300	500	100		2.2		
1969	8,900		89,000	100	200		0.2		
1970	2,747		44,830	236	72		1.8		
1971[4]									
1972	3,024		93,130	3,819	157		0.8	2.0	
1973									
1974									
1975	5197	409	72,750	1,313	159			0.1	1.4
1976									
1977	1700		34,000	384	2,700	1.4	0.6	2.0	
1978	165		18,480	593	212				0.1
1979	23	227	13,600		129				0.6
TOTAL	38041	636	587,777	7,600	4,398	1.4	5.6	4.1	2.1

[1]CM-243 and CM-244.

[2]Unidentified Alpha.

[3]Blanks indicate that no data was reported.

[4]No injections during 1971, 1973, 1974 and 1976.

Table 5. Curies in Liquid to Second Shale Fracture Facility

Year	SR-90	CS-137	CM-244	UN-ID AL[1]	Mixed F. P.[2]
1982	148,000	34,000	1,220	438	6,800
1983	453,000	43,300	4,510	1,290	6,500
1984	44,600	7,700	834	2,130	1,270
TOTAL	645,600	85,000	6,564	3,858	14,570

[1]UN-ID AL is unidentified alpha which consists of transuranics less CM-244.

[2]Mixed F.P. is unidentified beta-gamma emitters consisting primarily of mixed fission products.

TABLE 6. QUANTITIES TO SOLID WASTE (CURIES)

YEAR	TRU-U[1]	CS-137	H-3	OTHERS[2]	PU-239	SR-90	TH-232
1943-1976[3]	2.5E+03	1.7E+04	9.0E+04	1.1E+04	1.0E+02	4.5E+04	4.2E-01
1977	7.7E+00	1.2E+02	7.7E+01	3.3E+03	1.0E-02	1.8E+01	8.0E-03
1978	1.0E-01	2.3E+02	1.0E+03	2.3E+02	4.0E-03	2.1E+02	1.0E+00
1979	1.6E+00	3.5E+02	5.8E+02	3.9E+03	3.0E-01	1.1E+02	3.0E-02
1980	6.9E+00	1.4E+03	7.3E+01	5.4E+04	3.0E-02	2.4E+03	5.0E-01
1981	5.5E+00	2.1E+02	2.5E+01	1.1E+05	6.0E-02	1.2E+02	3.0E-04
1982	1.0E-01	6.9E+02	2.7E+03	3.4E+03	2.0E-03	5.1E+01	2.0E-02
1983	2.5E-01	9.4E+02	2.3E+03	1.8E+03	1.0E-01	1.7E+01	5.0E-01
1984	4.1E-01	1.2E+03	3.1E+02	9.0E+03	9.0E-03	1.6E+01	5.0E-02
1985	5.0E-01	1.4E+01	5.1E+02	1.0E+04	2.0E-02	4.0E-01	6.0E-04
TOTAL 1943-1985	2.5E+03	2.3E+04	9.8E+04	1.4E+05	1.0E+02	4.9E+04	2.6E+00

[1]Transuranics other than ^{239}Pu.

[2]Others consist of all beta gamma not specifically listed (includes total rare earths).

[3]Best estimate for given time period. Insufficient annual data.

Table 7. DOE-ORNL - 1958-1984

Total Uranium Contained in Solid Waste Buried on Site and Retrievable Storage

Fiscal Year	Enriched Uranium			^{233}U			Depleted U			Normal U	
	U(g)	^{235}U(g)	Ci	U(g)	^{233}U(g)	Ci	U(kg)	^{235}U(kg)	Ci	(kg)	Ci
1958											
1959				45	45	.43					
1960											
1961	8,245	5,995	0.33	4	4	.04					
1962	14,463	653	0.04	30	30	.28					
1963	2,899	242	0.01	1	1	0.01				224	0.15
1964	45,445	5,960	0.34	2,379	380	3.60				10	0.01
1965	19,136	904	0.05	12	11	0.10				45	0.03
1966	2,261	530	0.03	51	50	0.47				63	0.04
1967	4,023	974	0.06	2,204	1,808	17.14				51	0.03
1968	2,539	1,633	0.08	88	81	0.77				11	0.01
1969	1,964	696	0.04	433	402	3.81				52	0.04
1970	3,429	555	0.03	591	556	5.27	12,838	33	4.33	44	0.03
1971	34,103	13,238	0.68	680	670	6.35	118		0.04	34	0.02
1972	4,895	611	0.02	90	68	0.64	65		0.02	32	0.02
1973	3,727	559	0.03	306	293	2.78	39	1	0.01	4	0.003
1974	6,372	1,599	0.10	1,258	1,230	11.66	717	1	0.24	31	0.02
1975	6,921	983	0.05	1,373	1,341	12.71	1,163	3	0.39	425	0.30
1976	977	854	0.06	2,886	2,284	21.65	25		0.01	63	0.04
1977											
1978*	83,395	40,251	2.08	5,101	3,809	36.11	1,383		0.46	464	0.32
1979											
1980											
1981	280	203	0.01	599	579	5.49	906	3	0.31	6	0.004
1982	2,625	1,721	0.08	3		0.00	4		0.00	5	0.003
1983	1,045	318	0.02	6	6	0.06	163		0.05	6	0.004
1984	18,513	792	0.05	81	79	0.75	248		0.08	0	
Totals	267,257	79,271	4.2	18,221	13,727	130.	17,669	41	5.94	1,570	1.07

*Quantities are for the years 1977 through 1980. Detail by year is not available.

Table 8. Estimated Atmospheric Releases of Radioactivity at ORNL

Year	Radionuclide Discharge (Ci)				Unidentified Alpha
	I-131	H-3	Kr-85	Xe-133	
1984	0.10	33,400[a]	14,900	72,700	9.6E-08
1983	0.05	22,200	11,900	57,700	4.3E-06
1982	0.13	19,000	11,700	57,100	2.7E-06
1981	0.50	11,300	6,700	32,400	7.8E-08
1980	0.22	14,800	8,800	42,800	4.9E-06
1979	0.30	5,109	10,500	51,190	4.8E-06
1978	1.70	2,500	12,000	59,000	4.0E-06
1977	1.37	2,524	8,606	42,030	4.0E-06
1976	1.25	6,019	11,500	54,000	4.0E-06
1975	2.10	534	17,700	87,500	4.0E-06
1974	1.97	555	20,000	99,200	4.0E-06
1973	2.18	9,100	14,000	68,600	4.0E-06
1972	1.70	1,800	15,400	64,900	4.0E-06
1971	3.46	b	15000	71,000	b
1970	1.43[c]	b	15,000	75,000	b
1969	16.38	b	b	b	b
1968	10.38	b	b	b	b
1967	22.30	b	b	b	b
1966	15.79	b	b	b	b
1965	18.40	b	b	b	b
1964	84.50	b	b	b	b
1963	54.00	b	b	b	b
1962	121.20[d]	b	b	b	b
1961	42.00	b	b	b	b

[a]First tritium release estimate developed from monitoring data rather than from a calculation based on radionuclide inventory.

[b]No data.

[c]First estimate reflecting the effects of an upgraded charcoal filter system.

[d]First estimate based on in-stack sampling information.

APPENDIX A

APPENDIX A

ENVIRONMENTAL MONITORING DATA

Selected environmental data from annual ORNL and DOE (or predecessor organizations) Oak Ridge Operations environmental reports are shown in tabular form. Refer to the original reports for maps of sampling networks, for additional data, and for more details related to the data. A bibliography for all data is given at the end of the appendix.

In general, data tables which pertain to ORNL on-site data are from Applied Health Physics Annual Reports, and Industrial Safety and Applied Health Physics Annual reports for the appropriate year. Tables which pertain to off-site data are taken from the Environmental Monitoring Reports.

Throughout the history of ORNL operations, environmental radionuclide data have been influenced by world-wide fallout from nuclear weapons tests. In addition there is always natural radioactivity in the environment. Due to these and additional man-made sources, the origin of radionuclides measured in the environment is not always apparent. An indication of the origin of a radionuclide may sometimes be inferred by comparing local and remote monitoring results and reviewing reported discharges from known sources or events.

Table A.1. Air Monitoring Data Summary
Gross Alpha, Gross Beta, and I-131, 1961-1984
(E-6 pCi/cc)

Year	Gross Alpha		Gross Beta		Iodine-131	
	Local	Remote	Local	Remote	Local	Perimeter
1961			1.6	1.7		
1962			3.7	4.3		
1963			4.9	4.3		0.015
1964			1.3	1.1		0.03
1965			0.28	0.19		0.015
1966			0.17	0.11	0.23	0.014
1967			0.22	0.10	0.43	0.019
1968			0.29	0.16	0.13	0.013
1969			0.26	0.16	0.31	0.018
1970			0.33	0.23	0.03	0.01
1971			0.44	0.64	0.061	0.011
1972			0.13	0.084	0.037	0.011
1973			0.065	0.028	0.047	0.009
1974			0.10	0.084	0.029	0.008
1975			0.058	0.042	0.025	0.007
1976			0.046	0.026	0.026	0.008
1977	0.0019	0.0009	0.062	0.049	0.062	0.007
1978	0.0021	0.0011	0.089	0.076	0.031	0.008
1979	0.0021	0.0009	0.044	0.024	0.021	0.004
1980	0.0019	0.0011	0.052	0.029	0.003	0.001
1981	0.002	0.0011	0.089	0.070	0.003	0.001
1982	0.0022	0.0010	0.043	0.022	0.003	0.001
1983	0.0022	0.0010	0.051	0.024	0.005	0.001
1984	0.0026	0.0011	0.027	0.014	0.004	0.002

Table A.2. Water Monitoring Results
Clinch River - Below ORNL, 1961 - 1984
(pCi/l)

Year	Potable Water Sr-90	River Water (CRM 4.5, Center's Ferry)* Sr-90	Ce-144	Cs-137	Ru-103/ Ru-106	Co-60	Zr-95 Nb-95	H-3
1961	0.3	4.3	1.1	0.5	390	3.9		
1962	0.5	2.3	1.0	0.7	160	3.2		
1963		2.9	2.7	2.3	80	4.7	4.0	
1964	1.3	2.8	1.0	3.3	45	4.3	0.25	
1965	0.64	1.5	0.4	1.7	12	2.3	0.1	
1966	0.79	2.1	0.6	1.6	3.9	3.7	<0.1	
1967	0.80	1.2	0.2	1.5	0.6	1.2	0.1	
1968	0.35	1.5	0.4	1.4	0.7	2.7	0.2	
1969	0.43	1.1	0.2	1.9	1.4	3.5	0.4	
1970	0.51	1.1	0.3	1.9	0.5	1.0	0.2	2,200
1971	0.51	1.0	0.3	0.9	2.0	0.8	<0.1	1,850
1972	0.40	1.1		0.5	0.6			<1,620
1973	0.44	1.2		0.5	0.5			<1,530
1974	0.34	0.45		0.08	0.31			1,240
1975	0.60	0.31		0.05	0.19			1,100
1976	0.05	0.24		0.02	0.15			1,900
1977	0.12	0.27		0.5	0.19			1,000
1978	0.10	0.10		0.71	0.73			2,200
1979	0.46	0.33		0.02	0.11			1,590
1980	0.68	0.75		0.08	0.17			5,580
1981	1.5	1.4		0.12	0.03	0.1		1,640
1982	1.1	1.8		0.67		0.6		3,400
1983	0.51	2.0		0.28		0.2		3,800
1984	0.7	2.2		<0.54		<0.54		1,700

*Sample point deleted in 1980 - Gallaher data used for 1980 and following.

Table A.3. Water Monitoring Results, Clinch River - Above ORNL Discharge, 1961-1984

(pCi/l)

Year	Sr-90	Ce-144	Cs-137	Ru-103 Ru-106	Co-60	Zr-95 Nb-95	H-3
1961	0.7	0.7	0.5	5	0.4		
1962	1.6	1.4	0.2	7.8		4.2	
1963	1.3	2.0	0.2	9.2	0.1	2.6	
1964	1.2	0.8	0.4	3.1		0.05	
1965	0.6	0.3	0.3	2.4			
1966	0.9	0.3	0.3	0.6			
1967	0.4	0.1	<0.1	0.3		<0.01	
1968	0.4	0.2		0.6	4.4		
1969	0.8	0.3	0.9	0.8	3.8	0.4	
1970	0.5	0.2	0.4	0.4	0.3	0.2	1,400
1971	0.5	0.3	<0.1	1.2	0.3	<0.1	<1,000
1972	0.5	0.1	0.6				<1,000
1973	0.5	0.3	0.3				<1,080
1974	0.09	0.03	0.10				930
1975	0.14	0.05	0.09				820
1976	0.08	0.02	0.13				610
1977	0.15	0.0	0.10				494
1978	0.09	0.45	1.36		0.09		603
1979	0.10	0.01	0.05		0.01		646
1980	0.11		0.07	0.17	0.11		760
1981	1.30		0.003		0.20		1,420
1982	1.4		0.22		0.18		910
1983	0.93		0.088		0.08		12,000
1984	0.36		<0.37		<0.32		<4,100

Table A.4. Milk Results for Samples Collected Within 25 Miles of ORNL, 1961-1984 (pCi/liter)

Year	Local		Remote	
	Sr-90	I-131	Sr-90	I-131
1961	11	8		
1962	33	96	20	30
1963	43	13		
1964	20	11	23	
1965	19.8	7.4	20.4	
1966	26.1	9.0	24.4	
1967	28	17	25	6.4
1968	20	5.7	19	
1969	17.8	5.9	15.7	
1970	11.4	<10	9.3	<10
1971	11.4	<10	9.4	<10
1972	11	<11.4	8.6	<13.3
1973	8.1	<10	6.1	<10
1974	4.1	<0.49	2.7	<.65
1975	3.7	<0.56	3.2	<.47
1976	2.8	3.0	2.8	5.4
1977	3.4	<1.9	2.5	<1.1
1978	3.1	<0.76	2.5	<0.59
1979	2.5	<0.52	1.9	<0.45
1980	1.7	<0.45	1.6	<0.45
1981	1.6	<0.45	1.3	<0.45
1982	1.4	<0.45	1.4	<0.45
1983	1.2	<0.45	1.1	<0.45
1984	<1.6	<0.8	<1.2	<0.8

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BIBLIOGRAPHY

Data Year[a]	Report Number
Applied Health Physics Annual Reports	
1961	ORNL-3284
1962	ORNL-3490
1963	ORNL-3665
1964	ORNL-3820
1965	ORNL-3969
1966	ORNL-4146
1967	ORNL-4286
1968	ORNL-4423
1969	ORNL-4563
1970	ORNL-4690
1971	ORNL-4795
1972	ORNL-4894
1973	ORNL-4974
1974	ORNL-5055
1975	ORNL-5169
1976	ORNL-5310
Industrial Safety and Applied Health Physics Annual Reports	
1977	ORNL-5420
1978	ORNL-5543
1979	ORNL-5663
1980	ORNL-5821
1981	ORNL-5859
1982	ORNL-5962
Environmental and Occupational Safety Division Annual Progress Report	
1983	ORNL-6077

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BIBLIOGRAPHY (Cont.)

Data Year[a]	Report Number
Environmental Monitoring Report, United States Atomic Energy Commission, Oak Ridge Facilities	
1972	UCC-ND-244
1973	UCC-ND-280
Environmental Monitoring Report, Energy Research & Development Administration, Oak Ridge Facilities	
1974	UCC-ND-302
1975	Y/UB-4
1976	Y/UB-6
1977	Y/UB-8
Environmental Monitoring Report, United States Department of Energy, Oak Ridge Facilities	
1978	Y/UB-10
1979	Y/UB-13
1980	Y/UB-15
1981	Y/UB-16
1982	Y/UB-18
1983	Y/UB-19
1984	ORNL-6209

[a]The year of publication was always one year after data year.

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